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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of  
Hirokazu TANAKA, et al.

Group Art Unit: 1773  
Examiner: AHMED SHEEBA

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For: coating liquid for forming Hard Coat Film and Substrate Coated  
With Such a Film

Honorable Commissioner of Patents and Trademarks  
United States Patent and Trademark Office  
Washington, D. C. 20231

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DECLARATION UNDER 37 CFR 1.132

I, Hirokazu TANAKA, declare and state that:

1. I am a citizen of Japan, and residing at 13-2, Kitaminato-machi,  
Wakamatsu-ku, Kitakyushu-shi, Fukuoka, Japan.

In March 1969, I was graduated from Tokyo University of Science,  
and received a degree of Bachelor of Science.

Since April 1969, I have been an employee of Catalyst &  
Chemicals Industries Co., Ltd. Simultaneously, I had been  
assigned to Wakamatsu Plant and engaged in the development of  
novel inorganic materials, especially the production of the fine  
inorganic powders.

2. I am a co-inventor of the invention described in the

specification of the above-identified application.

3. The following Experimental Test was carried out in order to demonstrate the unobviousness of the present invention.

#### Experiment

##### Comparative Example A

0.04 part by weight, in term of  $\text{Fe}_2\text{O}_3$ , of ferric chloride and 99.96 parts by weight, in term s of  $\text{TiO}_2$ , of titanium tetrachloride were dissolved in pure water, thereby obtaining 10,000 parts by weight of an aqueous solution of mixture. 15% aqueous ammonia was slowly added to the aqueous solution of mixture until the pH thereof became 9.0, thereby obtaining a co-precipitate gel of iron oxide hydrate and titanium oxide hydrate.

The thus obtained co-precipitate gel was dehydrated and washed. 1150 part by weight of 35% hydrogen peroxide and 250 parts of the pure water were added to 1100 parts by weight of the co-precipitated gel, and heated to  $80^\circ\text{C}$ . As are result, a reddish-brown solution was obtained. This solution was diluted with pure water so that the concentration of iron and titanium oxides contained in the solution, in terms of  $(\text{TiO}_2 + \text{Fe}_2\text{O}_3)$ , became 1.0% by weight, and heated at  $200^\circ\text{C}$  for 9 hr in autoclave.

Thereafter, the water of the solution was replaced by methanol, and the mixture was concentrated until the concentration in terms of  $(\text{TiO}_2 + \text{Fe}_2\text{O}_3)$  became 20% by weight. Thus there was obtained a sol of composite metal oxide of iron oxide and titanium oxide (sol  $\text{E}_1$ ) having an average particle size of 11 nm and a weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  of 4/9996 (0.0004).

A coating liquid for forming a hard coat film was prepared in the same manner as in Example 1 of the present invention, except that the sol E<sub>1</sub> was used in place of the sol A<sub>1</sub>. Further, a hard coat film and a hard coat film having an antireflection coating were formed from coating liquid, and evaluated, in the same manner as in Example 1.

The results are shown in the following table A.

#### Comparative Example B

A sol of the composite metal oxide of iron oxide and titanium oxide having an average particle size of 11 nm (sol F<sub>1</sub>) was produced in the same manner as in Example 1 of the present invention, except that the amounts of ferric chloride and titanium tetrachloride were regulated so that Fe<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> (weight ratio) became of 7/993 (0.007).

A coating liquid for forming a hard coat film was prepared in the same manner as in Example 1 of the present invention, except that the sol F<sub>1</sub> was used in place of the sol A<sub>1</sub>. Further, a hard coat film and a hard coat film having an antireflection coating were formed from coating liquid, and evaluated, in the same manner as in Example 1.

The results are shown in the following table A

Table A

	sol		Photo chromis m	High refrati vity	Scuffi ng Resist ance	Appea rance	Dye affinit y	water resistance		Clou dine ss	Stability	
	Fe <sub>2</sub> O <sub>3</sub> /Ti O <sub>2</sub>	SiO <sub>2</sub> /(Fe 2O <sub>3</sub> +TiO 2)						colorin g	adhe renc e		25 days	45 days
Example 1	1/499		none	o	A	o	o	none	o	o	o	x
Comp.E x. 1	2/98		exhibited	o	A	o	o	occured	o	o	o	x
Example 2	1/499		none	o	A	o	o	none	o	o	o	Δ
Example 3	1/499	15/100	none	o	A	o	o	none	o	o	o	o
Example 4	1/999		none	o	A	o	o	none	o	o	o	x
Comp.E x. A	4/9996		exhibited	o	A	o	o	none	x	o	o	x
Comp.E x. B	7/993		exhibited	o	A	o	o	occured	o	o	o	x

Transmission of the hard coat films of Comparative Example A and B are lied intermediates between that of Example 1 and that of Comparative Example 1 in Fig 1 of the specification. But that of Comparative Example A and B are slightly lower than that of Example 1.

4. From the result of the above Experiment and based on my best knowledge and experience on the inorganic material development, I conclude as follows.

The hard coat film of Comparative Example A has photochromism and glows slightly blue at the irradiation of ultraviolet rays.

The hard coat film of Comparative Example B has photochromism and glows yellow at the irradiation of ultraviolet rays.

Furthermore, water resistance of the hard coat films of

Comparative Example A and B are insufficient.

Accordingly, the composite metal oxide having the limited weight ratio of the iron oxide to the titanium oxide of the present invention is specifically free from photochromism.

The undersigned declares further that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This 6 day of December 2002

  
Hirokazu TANAKA